

Translation

PATENT COOPERATION TREATY

PCT/EP2003/005952



# PCT

## INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 0000053647	FOR FURTHER ACTION	See Form PCT/IPEA/416
International application No. PCT/EP2003/005952	International filing date (day/month/year) 06 June 2003 (06.06.2003)	Priority date (day/month/year) 14 June 2002 (14.06.2002)
International Patent Classification (IPC) or national classification and IPC C08J 9/00		
Applicant BASF AKTIENGESELLSCHAFT		

1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 5 sheets, including this cover sheet.

3. This report is also accompanied by ANNEXES, comprising:

a. ☒ (sent to the applicant and to the International Bureau) a total of 3 sheets, as follows:

☐ sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).

☐ sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.

b. ☐ (sent to the International Bureau only) a total of (indicate type and number of electronic carrier(s)) \_\_\_\_\_, containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).

4. This report contains indications relating to the following items:

☒ Box No. I Basis of the report

☐ Box No. II Priority

☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

☐ Box No. IV Lack of unity of invention

☒ Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

☐ Box No. VI Certain documents cited

☐ Box No. VII Certain defects in the international application

☐ Box No. VIII Certain observations on the international application

Date of submission of the demand 02 December 2003 (02.12.2003)	Date of completion of this report 16 July 2004 (16.07.2004)
Name and mailing address of the IPEA/EP	Authorized officer
Facsimile No.	Telephone No.

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International Application No.

PCT/EP2003/005952

## Box No. I Basis of the report

1. With regard to the language, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.

- ☐ This report is based on translations from the original language into the following language \_\_\_\_\_, which is language of a translation furnished for the purpose of:
- ☐ international search (under Rules 12.3 and 23.1(b))
  - ☐ publication of the international application (under Rule 12.4)
  - ☐ international preliminary examination (under Rules 55.2 and/or 55.3)

2. With regard to the elements of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report)*:

- ☒ The international application as originally filed/furnished
- ☒ the description:
- pages \_\_\_\_\_ 1-10 \_\_\_\_\_, as originally filed/furnished
- pages\* \_\_\_\_\_ received by this Authority on \_\_\_\_\_
- pages\* \_\_\_\_\_ received by this Authority on \_\_\_\_\_
- ☒ the claims:
- pages \_\_\_\_\_, as originally filed/furnished
- pages\* \_\_\_\_\_, as amended (together with any statement) under Article 19
- pages\* 1-24 received by this Authority on 12 May 2004 (12.05.2004)
- pages\* \_\_\_\_\_ received by this Authority on \_\_\_\_\_
- ☒ the drawings:
- pages \_\_\_\_\_ 1/1 \_\_\_\_\_, as originally filed/furnished
- pages\* \_\_\_\_\_ received by this Authority on \_\_\_\_\_
- pages\* \_\_\_\_\_ received by this Authority on \_\_\_\_\_
- ☐ a sequence listing and/or any related table(s) – see Supplemental Box Relating to Sequence Listing.

3. ☐ The amendments have resulted in the cancellation of:

- ☐ the description, pages \_\_\_\_\_
- ☐ the claims, Nos. \_\_\_\_\_
- ☐ the drawings, sheets/figs \_\_\_\_\_
- ☐ the sequence listing (*specify*): \_\_\_\_\_
- ☐ any table(s) related to sequence listing (*specify*): \_\_\_\_\_

4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).

- ☐ the description, pages \_\_\_\_\_
- ☐ the claims, Nos. \_\_\_\_\_
- ☐ the drawings, sheets/figs \_\_\_\_\_
- ☐ the sequence listing (*specify*): \_\_\_\_\_
- ☐ any table(s) related to sequence listing (*specify*): \_\_\_\_\_

\* If item 4 applies, some or all of those sheets may be marked "superseded."

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No.

PCT/EP 03/05952

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement****1. Statement**

Novelty (N)	Claims	1-24	YES
	Claims		NO
Inventive step (IS)	Claims	1-24	YES
	Claims		NO
Industrial applicability (IA)	Claims	1-24	YES
	Claims		NO

**2. Citations and explanations**

Reference is made to the following document:

D1: WO 00/43442 A (EBERSTALLER ROMAN; SUNPOR KUNSTSTOFF  
GMBH (AT); ARDUINI SCHUSTER M) 2000-07-27

**1. Novelty (PCT Article 33(2))**

The subject matter of the present claims 1 to 24 is novel over the prior art as cited in the international search report. None of the international search report citations discloses a method for producing expandable styrene polymers with a molecular weight of more than 170,000 wherein a styrene polymer melt containing blowing agents is passed through a nozzle plate with bore holes no larger than 1.5 mm at a temperature of 140 to 300°C and then granulated.

**2. Inventive Step (PCT Article 33(3))**

The subject matter of the present claims 1 to 24 also involves an inventive step, since document D1, which can be considered the closest prior art, either alone or in combination with any other international search report citation, does not involve an inventive step for the following reasons:

Document D1 discloses a method for producing expandable styrene polymer particles with a molecular weight of 220,000 g/mol by passing a styrene polymer melt containing blowing agents through a nozzle with a bore hole no larger than 0.8 mm at a temperature of 120°C and then granulating it. The expandable styrene polymer granular particles produced in this way can furthermore contain fireproofing agents and pigments (cf. D1, example 1; page 4, lines 1-3; claims).

The method claimed in the present claims differs from the method known from D1 in that the styrene polymer melt is passed through the nozzle at a temperature of 140 to 300°C.

In light of the teaching of D1, the problem addressed by the present application is that of providing a method for producing expandable styrene polymer granulates that have small granulate sizes and a uniform size distribution of the granulates and that can easily be foamed into foam materials with a homogenous structure and a high cell count.

The applicant has demonstrated in numerous examples that the problem of interest can be solved by the claimed method using an increased extruder nozzle temperature.

Since document D1 expressly states that the temperature of the extruder nozzle should be cooled to 120°C in order to prevent premature foaming, this document does not suggest the claimed method. Therefore, an inventive step can be acknowledged.

We claim:

1. A process for the preparation of expandable styrene polymers  
5 having a molecular weight  $M_w$  of greater than 170,000 g/mol, which comprises conveying a blowing agent-containing styrene polymer melt having a temperature in the range from 140 to 300°C through a die plate with holes whose diameter at the die exit is at most 1.5 mm, and subsequently granulating the extrudate. {  
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2. A process as claimed in claim 1, wherein the expandable styrene polymer has a molecular weight in the range from 190,000 to 400,000 g/mol.  
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3. A process as claimed in claim 1 or 2, wherein the expandable styrene polymer has a molecular weight distribution having a polydispersity  $M_w/M_n$  of at most 3.5.
- 20 4. A process as claimed in any one of claims 1 to 3, wherein the styrene polymer employed is transparent polystyrene (GPPS), high-impact polystyrene (HIPS), an acrylonitrile-butadiene-styrene polymer (ABS), styrene-acrylonitrile (SAN) or a mixture thereof or with  
25 polyphenylene ether (PPE).
5. A process as claimed in any one of claims 1 to 4, wherein the blowing agent-containing styrene polymer melt comprises, in homogeneous distribution, from 2 to 10% by weight of one or  
30 more blowing agents selected from the group consisting of aliphatic hydrocarbons having from 2 to 7 carbon atoms, alcohols, ketones, ethers or halogenated hydrocarbons.
6. A process as claimed in any one of claims 1 to 5, wherein the  
35 blowing agent-containing styrene polymer melt comprises plasticizers, such as mineral oils, oligomeric styrene polymers and phthalates, in proportions in the range from 0.05 to 10% by weight, based on the styrene polymer.
- 40 7. A process as claimed in one or more of claims 1 to 6, wherein the blowing agent-containing styrene polymer melt is conveyed through the die plate at a temperature in the range from 160 to 240°C.

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8. A process as claimed in any one of claims 1 to 7, wherein the die plate is heated at least to the temperature of the blowing agent-containing polystyrene melt.
- 5 9. A process as claimed in any one of claims 1 to 8, wherein the diameter (D) of the die holes at the die exit is in the range from 0.2 to 1.2 mm.
- 10 10. A process as claimed in any one of claims 1 to 9, wherein the die plate has holes having an L/D ratio (length (L) of the die zone, whose diameter corresponds at most to the diameter at the die exit, to the diameter (D) at the die exit) of at least 2.
- 15 11. A process as claimed in any one of claims 1 to 10, wherein the diameter (E) of the holes at the die entrance of the die plate is at least twice as great as the diameter (D) at the die exit.
- 20 12. A process as claimed in any one of claims 1 to 11, wherein the die plate has holes having a conical inlet with an inlet angle  $\alpha$  of less than  $180^\circ$ .
- 25 13. A process as claimed in any one of claims 1 to 12, wherein the die plate has holes having a conical outlet with an outlet angle  $\beta$  of less than  $90^\circ$ .
- 30 14. A process as claimed in any one of claims 1 to 13, wherein the die plate has holes having different exit diameters (D).
- 35 15. A process as claimed in any one of claims 1 to 14, wherein the blowing agent-containing styrene polymer melt comprises 0.05 to 1.5% by weight of water, based on the styrene polymer.
- 40 16. A process for the preparation of expandable styrene polymers having a molecular weight Mw of greater than 170,000 g/mol, comprising the following steps:
- 45 a) polymerization of styrene monomer and, if desired, copolymerizable monomers,
- b) degassing of the resultant styrene polymer melt,

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- c) mixing of the blowing agent and, if desired, additives into the styrene polymer melt by means of static or dynamic mixers at a temperature of at least 150°C,
- 5 d) cooling of the blowing agent-containing styrene polymer melt to a temperature of at least 120°C,
- e) discharge through a die plate having holes whose diameter at the die exit is at most 1.5 mm, and
- 10 f) granulation of the blowing agent-containing melt.
- 17. A process as claimed in claim 15, wherein step f) is carried out directly behind the die plate under water at a pressure
- 15 in the range from 1 to 10 bar.
- 18. An expandable styrene polymer (EPS) obtainable by the process as claimed in claim 16, which comprises at most 500 ppm of styrene monomers.
- 20 19. An expandable styrene polymer (EPS) having a molecular weight  $M_w$  of more than 170,000 g/mol, which comprises from 0.05 to 1.5% by weight of internal water.
- 25 20. An expandable styrene polymer (EPS) as claimed in claim 19, wherein at least 90% of the internal water is present in the form of internal water droplets having a diameter in the range from 0.5 to 15  $\mu$ m.
- 30 21. An expandable styrene polymer (EPS) as claimed in claim 19 or 20, whose expansion capability  $\alpha$  is at most 125.
- 22. An expandable styrene polymer (EPS) as claimed in any of claims 18 to 21, which takes the form of granules having a
- 35 diameter of from 0.4 to 1.8 mm.
- 23. An expandable granulated styrene polymer (EPS) as claimed in claim 22, which has a bulk density of at most 700 g/l.
- 40 24. An expandable styrene polymer (EPS) as claimed in any of claims 18 to 22, which comprises from 0.01 to 30% by weight of pigments.

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